Experimental Quantification of Many-Body Interactions through the Alignment Measurements of Satellite States

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Excited ionic states produced during photoionization are referred to as satellite states due to their inherent weaker transition rate compared to the main photoline where the residual ion is unexcited. The observation of the fluorescence from these ionic states provides a sensitive method in the study of many-body interactions since measurements can be obtained at threshold with high resolution and efficiency [1]. Polarization analysis of this fluorescence allows one to determine magnetic-substate partial cross sections and the relative strength of the radial matrix elements for production of photoelectron partial waves [2]. We will further show that a measure of many-body interactions can be directly quantified from such a polarization analysis.

The satellite states of argon display a widely varying angular momentum content, the total angular momentum quantum number varying from $J = 1/2$ to as high as 9/2 [3]. Since the ionizing photon brings only one unit of angular momentum, this implies that there is a significant interaction between the residual atomic electrons and the continuum photoelectron in order to redistribute such an angular momentum content. With no hyperfine-structure, and its fine-structure resolvable by fluorescence, argon provides a convenient atomic system to explore and quantify the partitioning in such a redistribution. A quantitative measure of this angular momentum sharing can be gleaned from the determination of the total alignment of the excited ionic state since this atomic parameter, obtainable from a measurement of the linear polarization of the fluorescent radiation, is one measure of the distribution of the magnetic-substate population [2].

In the measurements reported here, an atomic beam of argon has been photoionized in the energy range from 35.4 to 36.6 *eV* with a resolution of 2 to 3 *meV*. This high resolution without a prohibitive loss of flux was achieved with the spherical-grating monochromator on the 10 *cm* undulator of ALS beam-line 9.0.1. The linearly polarized radiation [4] was made to intersect an effusive beam of argon with the linear polarization axis aligned along the atomic beam propagation axis. Fluorescence was observed orthogonal to this collision plane as well as along two collection axes within this plane: at 45^o and 60^o off axis from the quantization axis set by the polarization of the ionizing radiation. Each fluorescence collection system consisted of a photomultiplier coupled to the interaction region by a $f/1.9$ lens along with a linear polarizer and 0.3 *nm* bandwidth interference filter. By alternating the alignment of the linear polarizer, the fluorescent intensity parallel ($I_\parallel$) and perpendicular ($I_\perp$) to the linear polarization axis of the synchrotron radiation were measured at a fixed ionizing photon energy. Each individual intensity was normalized to the ionizing photon flux monitored by the displacement current from a biased aluminum surface intercepting the synchrotron beam as it exited the interaction region. Repeating the measurement upon incrementing the ionizing photon energy allowed the total intensity $I_t = I_\parallel + I_\perp$ and linear polarization $P = (I_\parallel - I_\perp)/I_t$ of a fine-structure resolved, satellite state fluorescence to be determined as a function of the ionizing photon energy.

Fig. 1 gives the total intensity spectra for four Ar^+ $4p \rightarrow 4s$ fluorescent transitions. The spectra shown is in the immediate vicinity of the threshold for the excitation of these $4p$ satellite states. As evidenced in Fig. 1, the onsets for these fine-structure resolved thresholds were abrupt with only the dark counts for the photomultiplier tubes registered prior to threshold. The ionizing photon energy was calibrated using the photoelectron yield for the Ar $3s \rightarrow np$ window resonances [5] along with six thresholds for similar satellite state fluorescence [2].

Overlain in the figures are predictions for Rydberg series which account for most of the structure of the spectra. Parity and angular momentum conservation restrict the orbital angular momentum of the Rydberg electron to only these values for the indicated excited ionic cores. The series predictions are based on previously accepted [3,5,6] quantum defects for argon along with the well-established [7] energy levels for the excited ionic cores.

Directly proportional to the expectation value of the zero component of the electric quadrupole tensor, the total alignment parameter $A_0(J)$, which is defined [8] as

$$A_0(J) \equiv \frac{\langle 3J_z^2 - J^2 \rangle}{J(J+1)} = \frac{\sum_{M_J} \sigma(J, M_J)\left[3M_J^2 - J(J+1)\right]}{J(J+1)\sum_{M_J} \sigma(J, M_J)} \qquad (1)$$

can be obtained from the measured linear polarization of each satellite state fluorescence [2]. The derived alignment spectra are shown in Fig. 2. Even though we determine the total alignment parameter from our polarization measurement of the $3p^4 4p$ satellite state fluorescence, we can *further quantify the alignment of both the* $4p$ *valence electron as well as that of the* $3p^4$ *subshell*. Decoupling the ionic total angular momentum into its orbital L and spin S components provides the relationship between the partial cross-sections for the orbital and total components:

$$\sigma(L, M_L) = \sum_{M_J} \langle L\, M_L\, S\, M_S | J\, M_J \rangle^2 \ \sigma(J, M_J)\,. \qquad (2)$$

Substituting (2) into an expression comparable to (1) for the alignment of the orbital angular momentum yields a relationship between the alignment parameter for the orbital and total angular momentum of the satellite state. This decomposition can be carried one step further by decoupling the total orbital angular momentum of the ion L into its constituent $4p$ electron orbital angular momentum ℓ and orbital angular momentum L_c for the $3p^4[^3\mathrm{P}]$ subshell. This decoupling allows the alignment parameter $A_0(L_c)$ for the orbital motion of the $3p^4$ subshell of each excited ionic state, as well as that for the $4p$ valence electron $A_0(\ell)$, to be written in terms of the measured alignment parameters $A_0(J)$ presented in Fig. 2. For example, for the $^2\mathrm{D}_{3/2}$ satellite state:

$$A_0(L_c) = A_0(\ell) = \tfrac{1}{2} A_0(L) = \tfrac{7}{16} A_0(J)\Big|_{[^3P]4p\,^2D_{3/2}}\,. \qquad (3)$$

Thus, for the autoionizing resonances in which the ionic core is isotropic, such as the following

$$\left\{\begin{matrix} 3p^4[^3P]\,4p\ \ ^2S\ ns,n'd \\ 3p^4[^1S]\,4s\ \ ^2S\ np \end{matrix}\right\} \xrightarrow{\text{autoionization}} 3p^4[^3P]\,4p \left\{\begin{matrix} ^2D_{3/2,5/2} \\ ^2P_{3/2} \end{matrix}\right\} + \varepsilon s,d$$

our alignment measurements of the $3p^4\,[^3\mathrm{P}]\,4p\ ^2\mathrm{D}_{3/2,5/2}$ and $^2\mathrm{P}_{3/2}$ satellite states provide an *experimentally accessible, direct quantitative measure of the many-body interaction* between the autoionizing $n\ell$ Rydberg electron with the $3p^4$ subshell and the $4p$ valence electron.

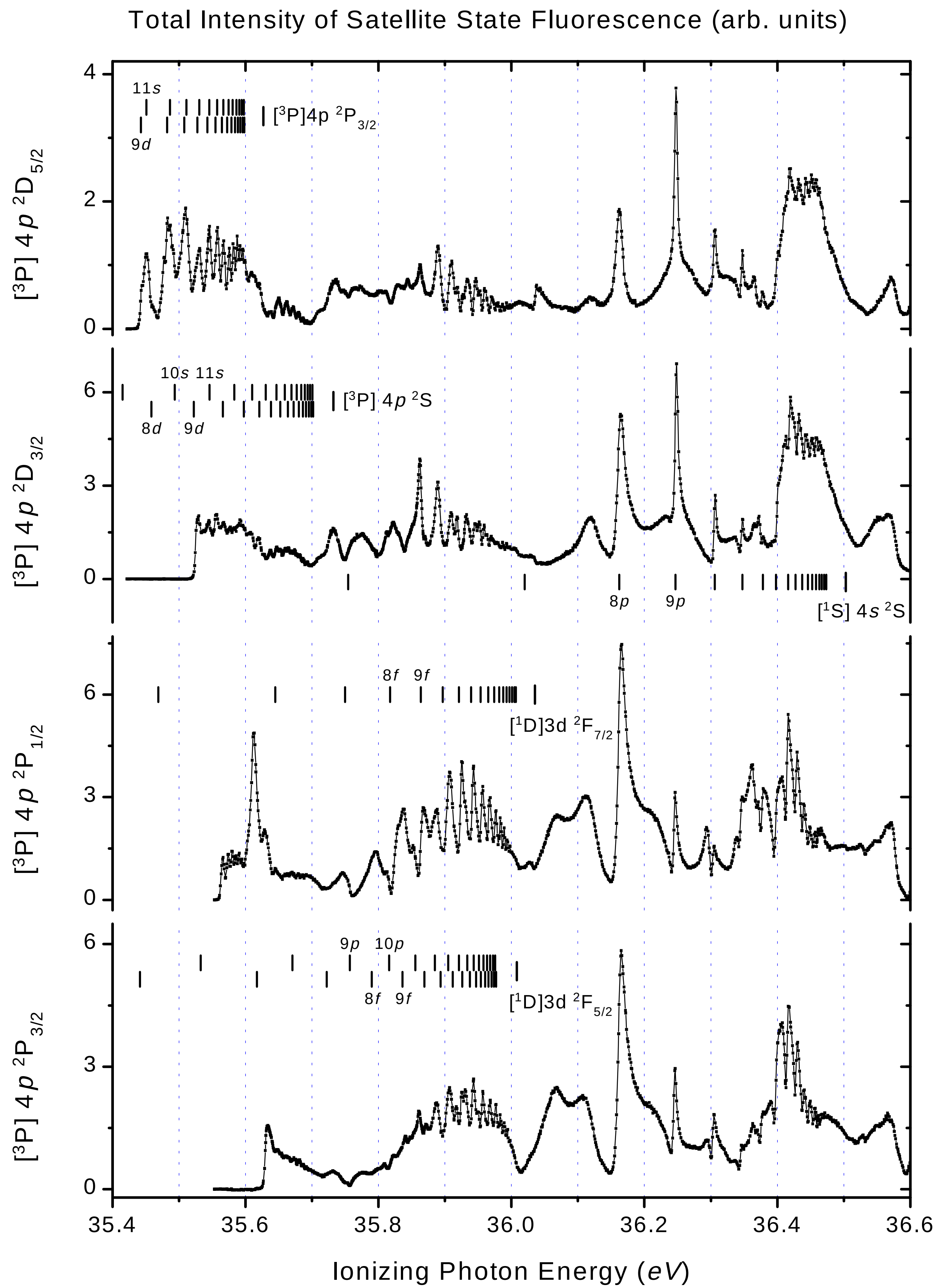

Figure 1. Total intensity of Ar^+ $3p^4$ [^{3}P] 4p ^{2}P and ^{2}D fine-structured resolved satellite state fluorescence in the near-threshold region.

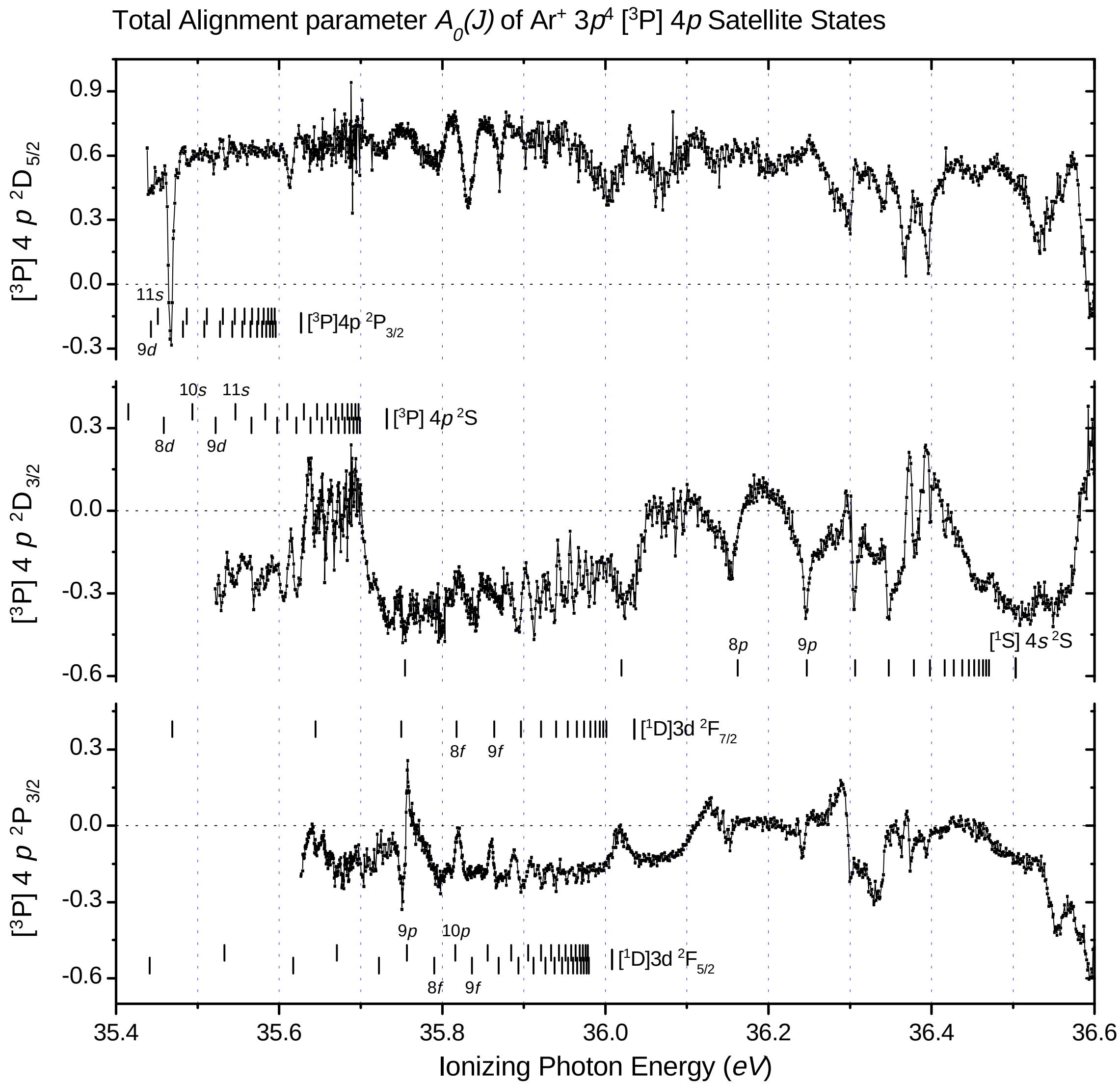

Figure 2. Alignment of the Ar^+ $3p^4$ $[^3P]$ $4p$ $^2P_{3/2}$ and $^2D_{3/2,\,5/2}$ satellite states in the near-threshold region.

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